FEG-STEM as a Tool for Nuclear Fuels and Reactor Materials Analysis

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Tecnai F-30 S-Twin FEG-STEM

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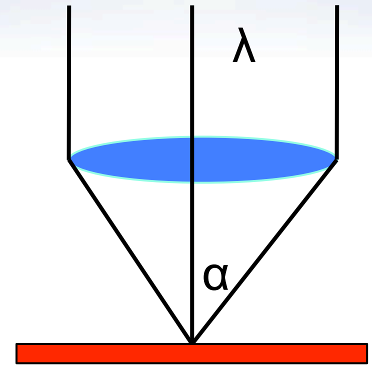
- ***EDS (Energy Dispersive Specstroscopy): High Spatial Resolution Chemical Analysis***
- ***Gatan Tridiem 863 P GIF: Energy Filtered TEM and STEM (EFTEM and EFSTEM, Electron Energy Loss Spectroscopy (EELS) : Chemical Analysis and Bonding States***
- ***BF/DF (Bright/Dark Field) and HAADF (High Angle Annular Dark-Field) STEM: Imaging and Z- Contrast respectively***
- ***Tomography Holder: 3 D Image Reconstruction***
- ***TEM Resolution: 2.0 Å***
- ***STEM HAADF Resolution: 1.9 Å***

Why use electrons for microscopy?

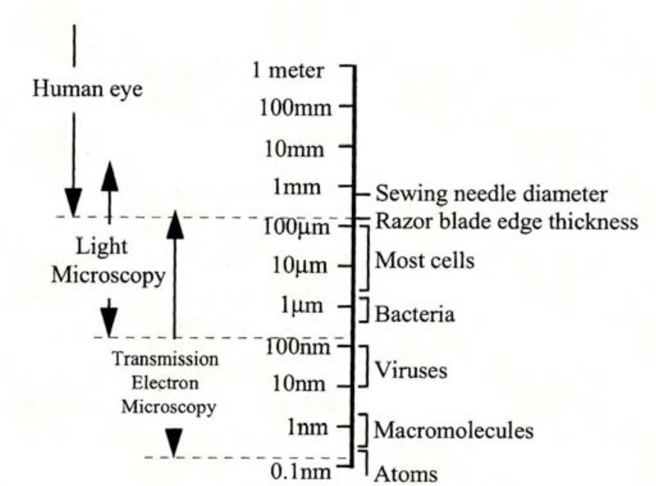
Abbe Theory of Resolution

$$d_d = 0.61\lambda/n \sin\alpha$$

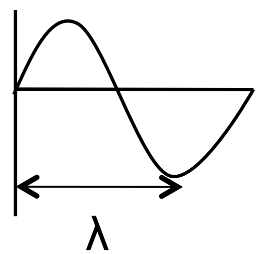
n = lens index of refraction
α = illumination semi-angle
λ = wavelength



Light microscope d_d ~ 200 nm
Electron microscope d_d on the order of angstroms



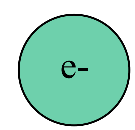
Wave



$\lambda = h/mv$

H: Plank's constant

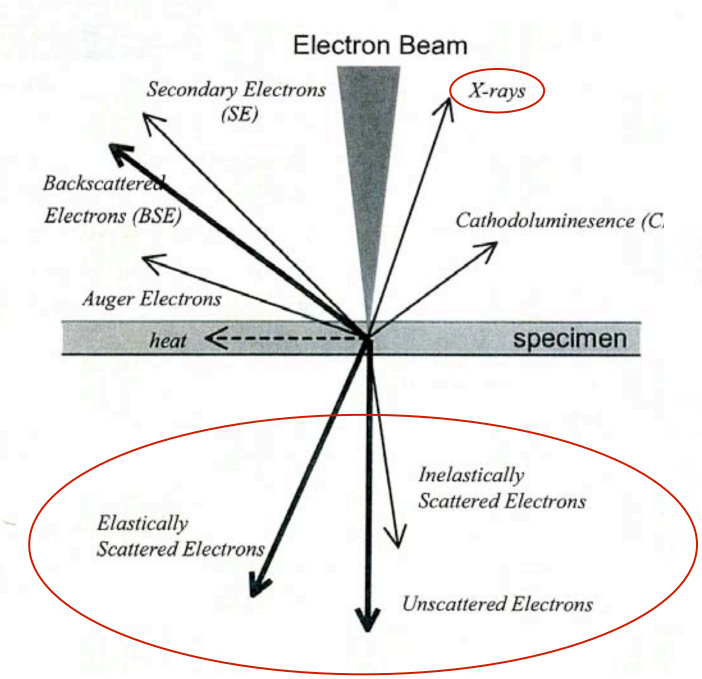
Particle

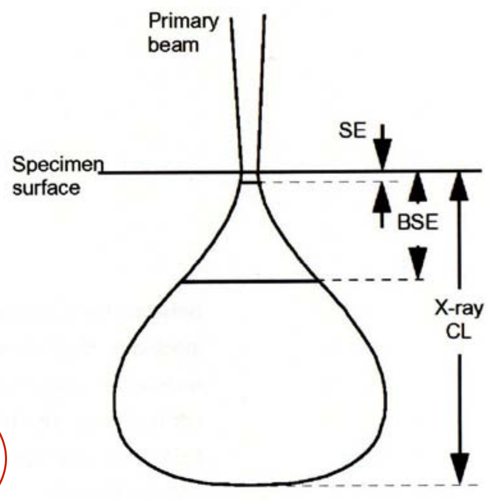


Mass m
Velocity v

- Charged particle nature of electron exploited using electromagnetic lenses to focus the beam of electrons
- Contrast, diffraction and microscope resolution are dictated by the wave nature of the electron

Electron-Specimen Interactions



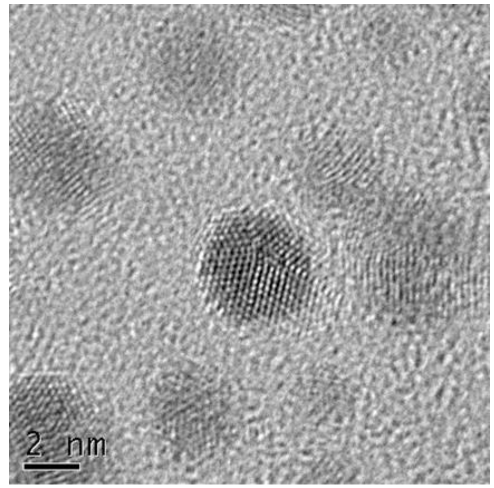


Elastic – change direction, not velocity (no energy loss)
Inelastic – change velocity (loses energy)

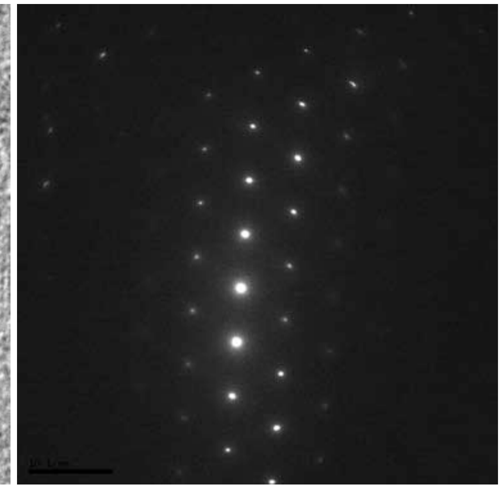
- Multiple signals are generated from the interaction of an electron beam with a material.
- These signals can be analyzed using various detectors, with each signal providing important information about the nature of the sample.
- Typical signals detected in FEG-STEM analysis include X-Rays (EDS), inelastically scattered (EELS, EFTEM), elastically scattered and unscattered electrons (diffraction, imaging) .
- The scattering of electrons in mater leads to broadening of the electron beam.
- Thin samples used in TEM (on the order of 50 – 100 nm) reduce this broadening which is critical for high spatial resolution analysis.

FEG-STEM in Radiation Materials Science

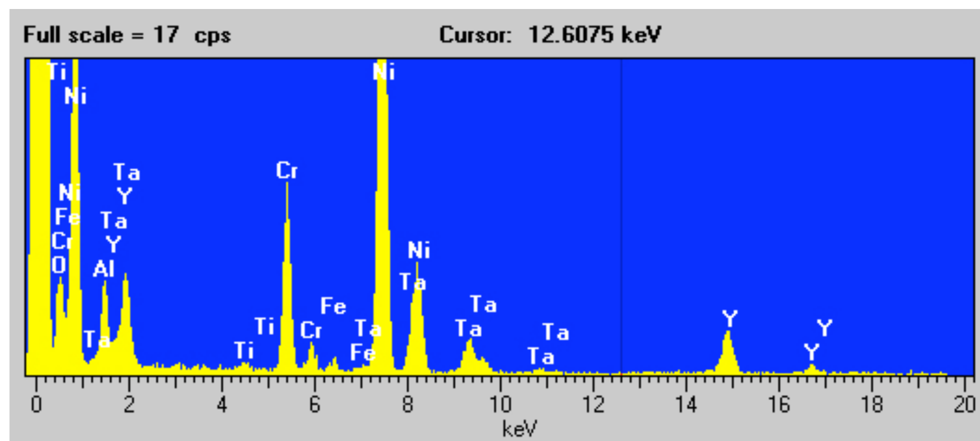
The TEM allows us to image the material microstructures at atomic resolution, determine crystal structure (atomic arrangements) and analyze chemical composition with a spatial resolution on the order of nanometers or in some cases angstroms. As such, it is ideally suited for characterizing radiation damage processes in fuels and materials that have there genesis in atomic level disturbances induced by high-energy particle interactions.



High Resolution Image



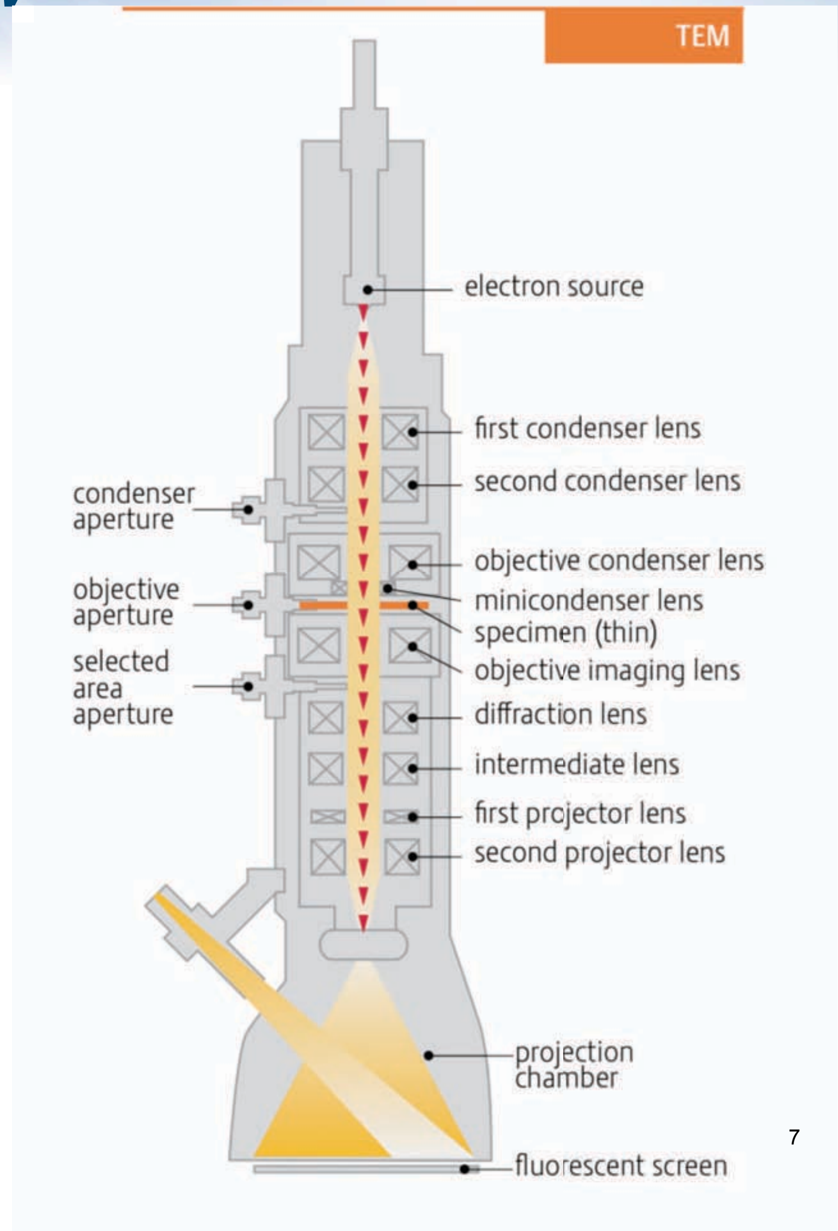
Selected Area Diffraction Pattern



EDS spectrum

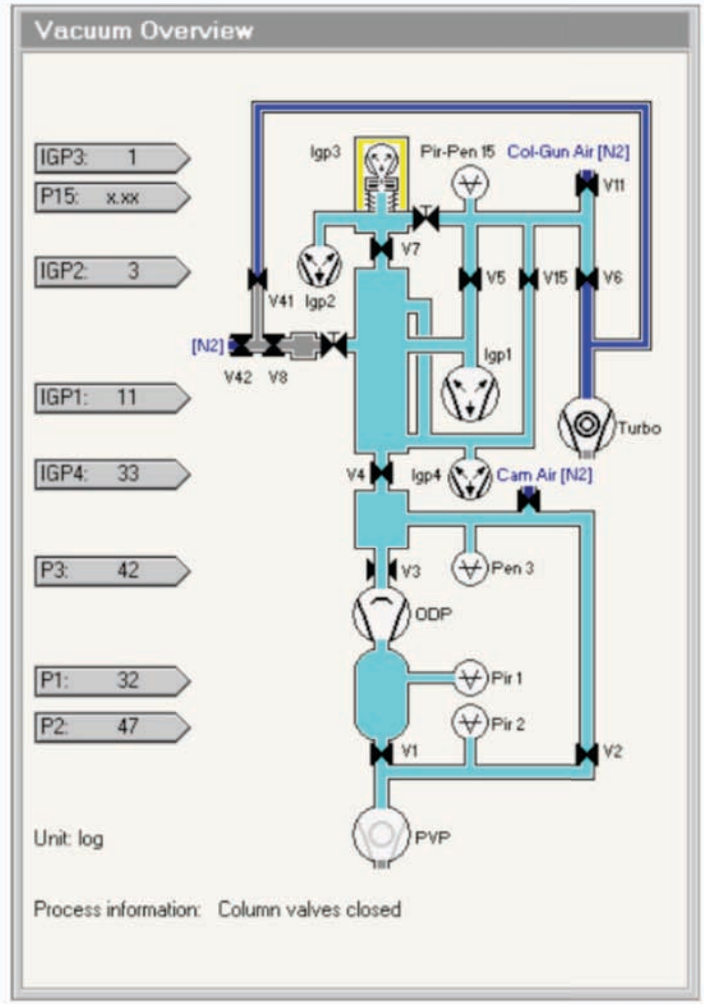
FEG-STEM System Overview

- Electron Source
  - Generates electrons that are accelerated by the high tension (voltage)
  - Can be Thermionic (W or LaB_6) or FEG
- Illumination System (Condenser)
  - Electron probe forming system allows control of probe intensity, diameter and convergence parameters.
  - The F-30 has two condenser lenses each having an adjustable aperture.
- Objective Lens
  - Produces the first intermediate image of the object (specimen).
  - Most critical lens in terms of aberrations/defects.
  - F-30 has twin-lens system with an adjustable aperture located in the back focal plan of the lens.
- Imaging Lens System
  - Magnifies image produced by objective lens.
  - Consists of 4 lenses in the F-30 and an adjustable selected area aperture.
- Projection chamber
  - Photo luminescent (phosphor) screen, image recording devices.



Vacuum System

- Reasons for ultra-high vacuum system
  - Electron gun needs high vacuum to maintain voltage stability and increase emitter lifetime (bad vacuum leads to etching of the W filament).
  - Collisions of electrons with gas molecules in column will degrade image quality.
  - Prevention of contamination on sample and internal surfaces.



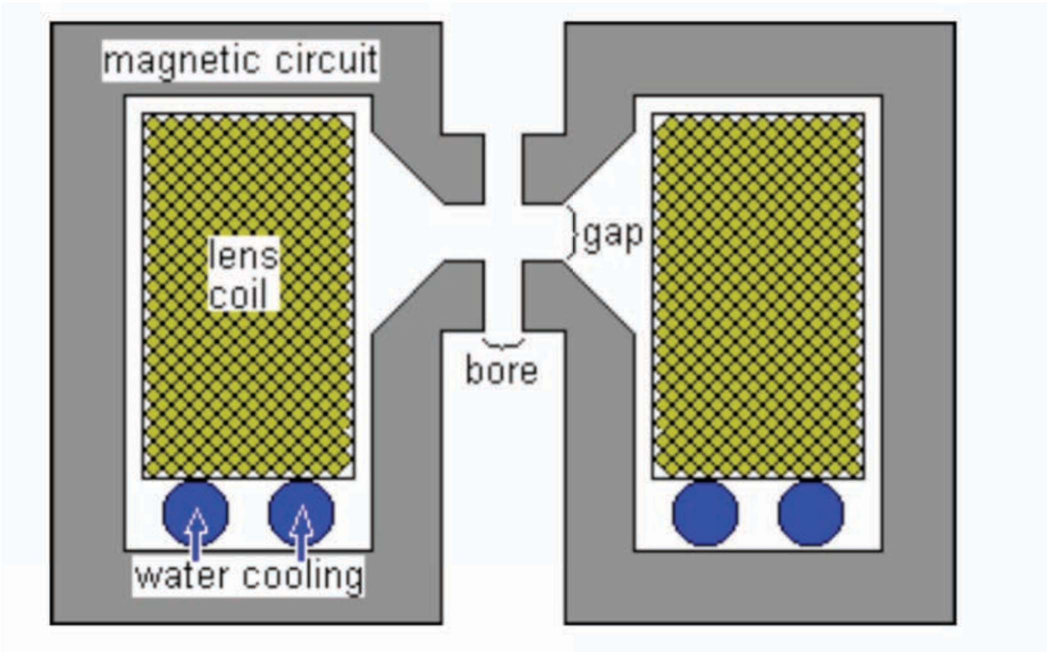
- Vacuum broken into multiple systems ranging from atmosphere to very high vacuum (10^{-8} Pa for FEG) as no individual pump is designed to handle the whole range of required vacuum.
  - Vacuum system for gun and column is separate from camera chamber.
  - Differential pumping apertures (200 μm in diameter) keep high vacuum areas isolated from lower vacuum areas.

- Vacuum pumps found on F-30
  - PVP (Rotary Pump) and ODP (oil diffusion pump)

    Buffer Tank and Camera Chamber
  - TMP (turbo-molecular pump) and IGP (1-4) (Ion Getter Pump)

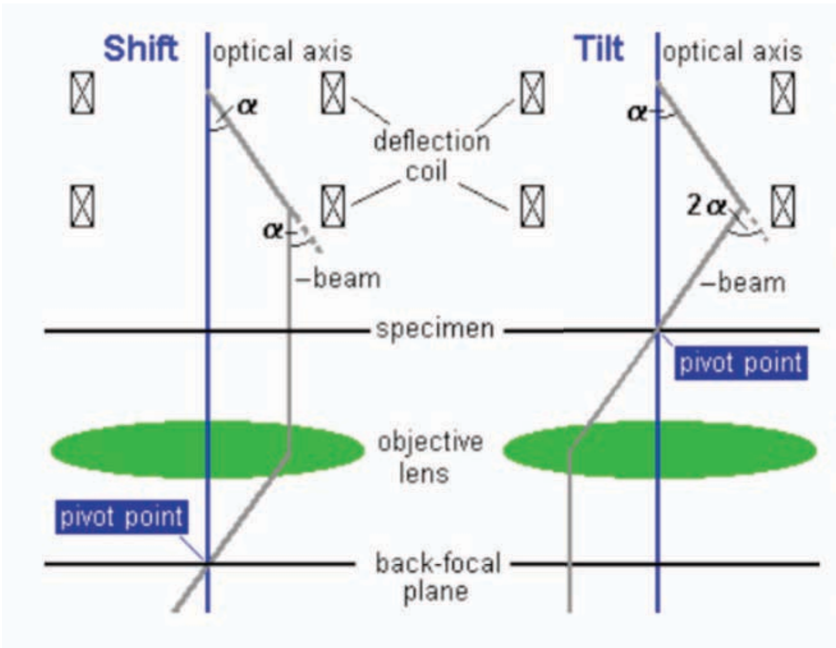
    Column and Gun

Electromagnetic Lenses

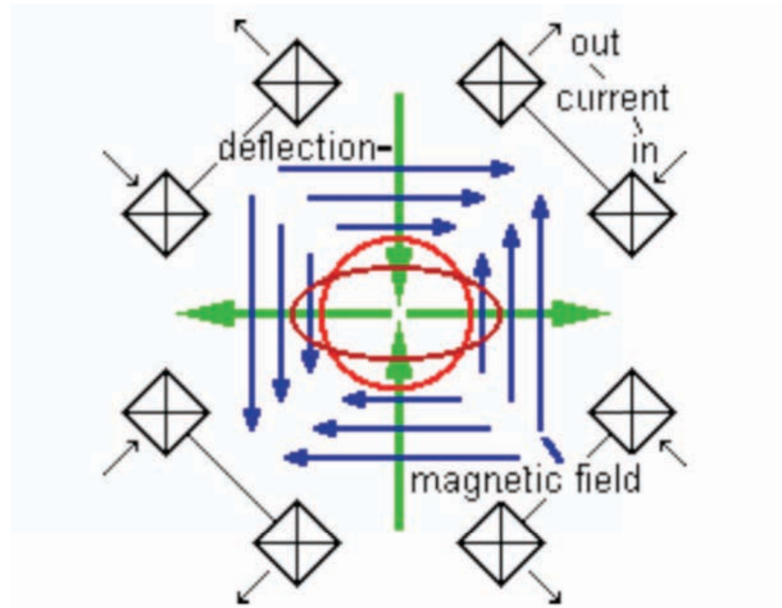


- Electrons are deflected in the magnetic field created by the lens.
- Outer soft iron core.
- Copper lens coils with water cooling to dissipate coil heating.
- Pole pieces shaped to optimize symmetry of magnetic field.
- Changing current through the lens coil changes magnetic field surrounding beam path.
- Electrons travel in a spiral path down the optical axis due to the magnetic forces acting on them.
- Spiraling of the electrons leads rotation of the final image with respect to the object.

Deflectors and Stigmators

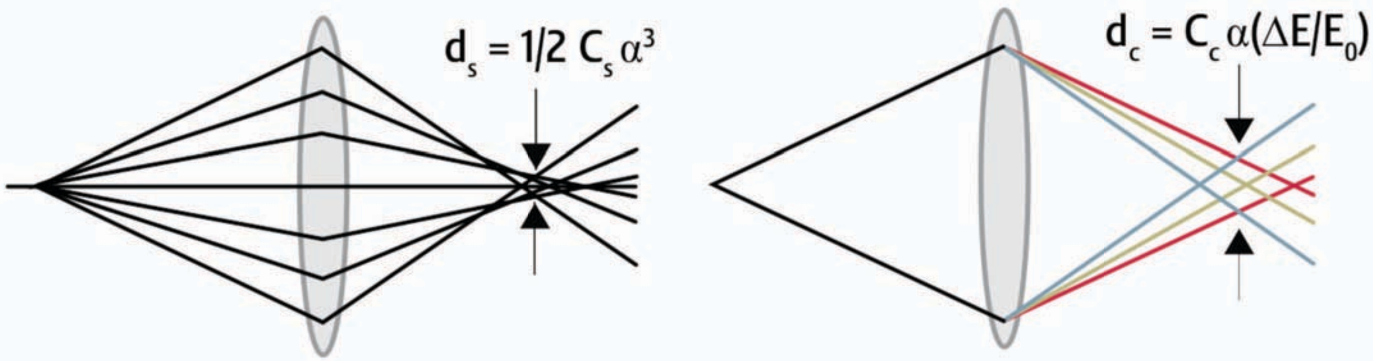


- For optimum resolution and probe intensity, beam should be parallel to and aligned with optical axis.
- Deflectors in column allow shifting and tilting of the beam to compensate for misalignment.
- Each lens system has a set of deflectors.



- Lens defect caused by asymmetry in the magnetic field of the lens.
- Image of a point is spread out in two dimensions.
- Sample dependent and can be influenced by sample (magnetism, contamination).
- Stigmators permit reshaping of the magnetic field.

Aberrations

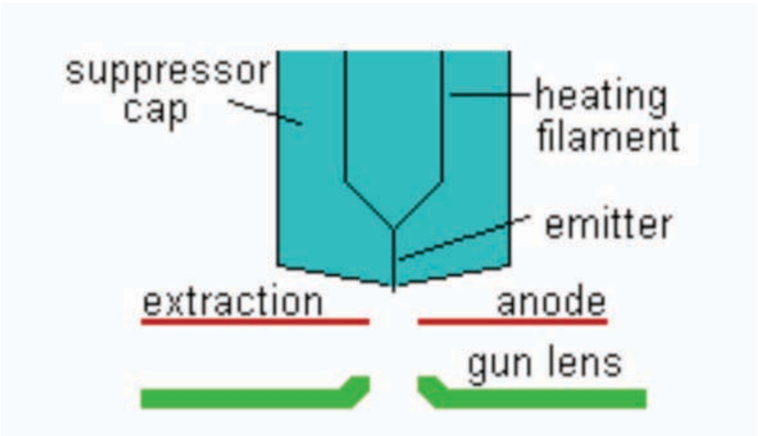


Spherical Aberration: Incident beams from the same point source do not focus to a point (impacted by lens design).

Chromatic Aberration: Waves of different energies converge at different focal plans (improves with stable electron source).

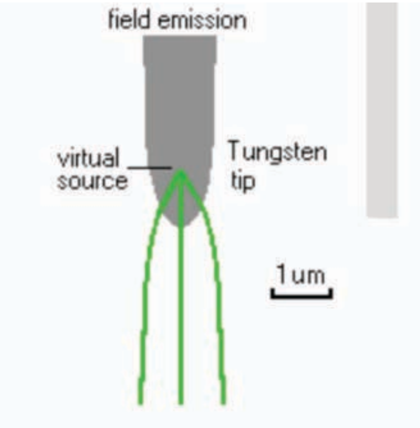
Aberrations limit resolution and probe intensity to significantly lower than theoretically attainable. Aberration corrected TEMs can improve resolution on the order of a factor of 2-3 (depending on microscope) and probe intensity by nearly an order of magnitude.

Field Emission Gun (Electron Source)



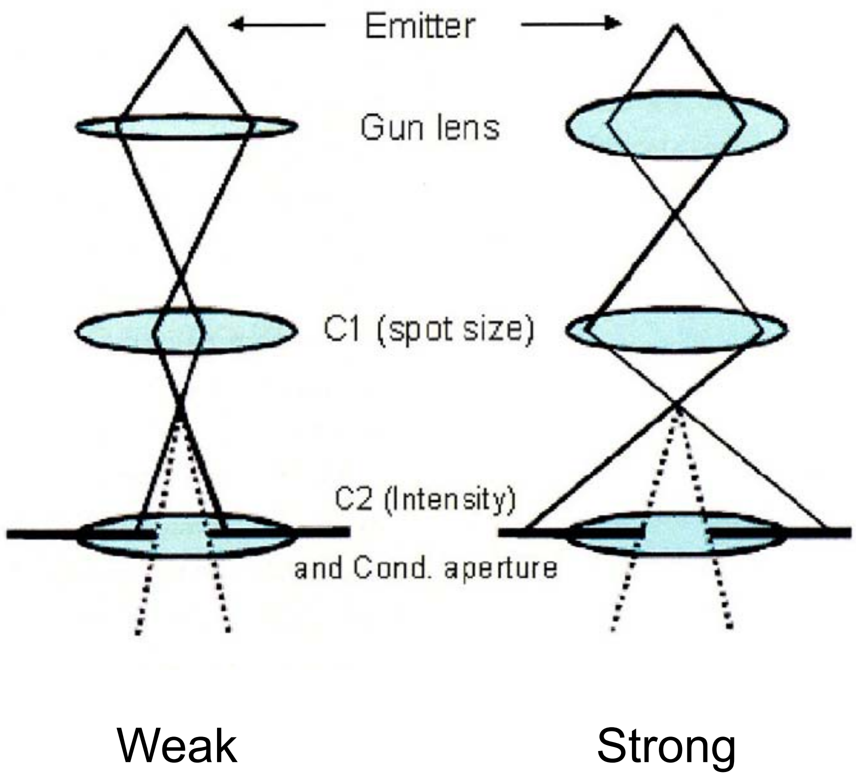
	W	LaB_6	FEG (Schottky)
Maximum Current (nA)	1000	500	300
Normalised Brightness (-)	1	10-30	2500
Energy spread (eV)	3-4	1.5-3	0.6-1.2
Source spotsize	30-100 µm	5-50 µm	15-30 nm
Required Vacuum (Pa)	10^{-3}	10^{-5}	10^{-7}
Temperature (K)	2700	2000	1800
Life time (hr)	60-200	1000	>2000
Normalised Price (-)	1	10	100

User controls extraction voltage (typically 3.5 to 4.5 kV) and gun lens setting. Emission current should be kept below 150 µA.



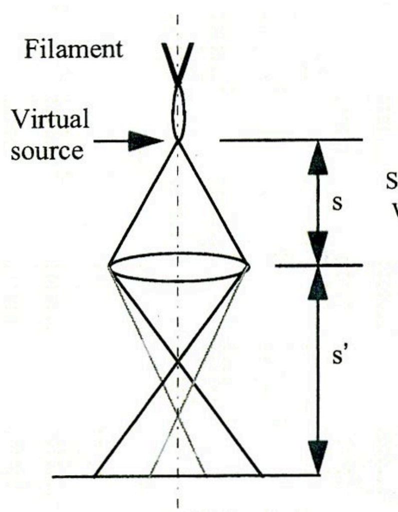
- Tungsten (100) Tip
  - F30 has a Schottky FEG
    - ZrO_2 coated tip (reduces work function)
    - Sharp tip (1 µm) leads to field of 10^7 V/cm allowing e^- to “tunnel” through energy barrier.
- Electrons originate from the center of the tip
  - Yields smaller apparent (virtual) source
  - Gives high brightness and sharp angular current distribution

Importance of Gun Lens

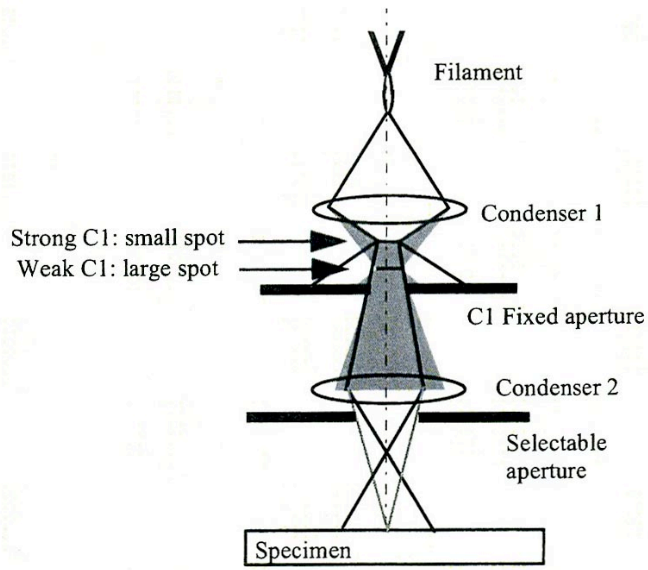


- Converges electron beam in column.
- Weakly excited (gun lens setting 1-3)
  - Crossover lower in the column
  - Higher total current
  - More aberrations
  - Good for TEM, HR-TEM and EFTEM TEM-EDS.
- Strongly excited (gun lens setting 4-9)
  - Crossover higher in column
  - Lower current, higher current density
  - Aberrations are minimized
  - Small probes can be formed
  - Good for STEM, EFSTEM and STEM-EDS.

Illumination System



Single Condenser



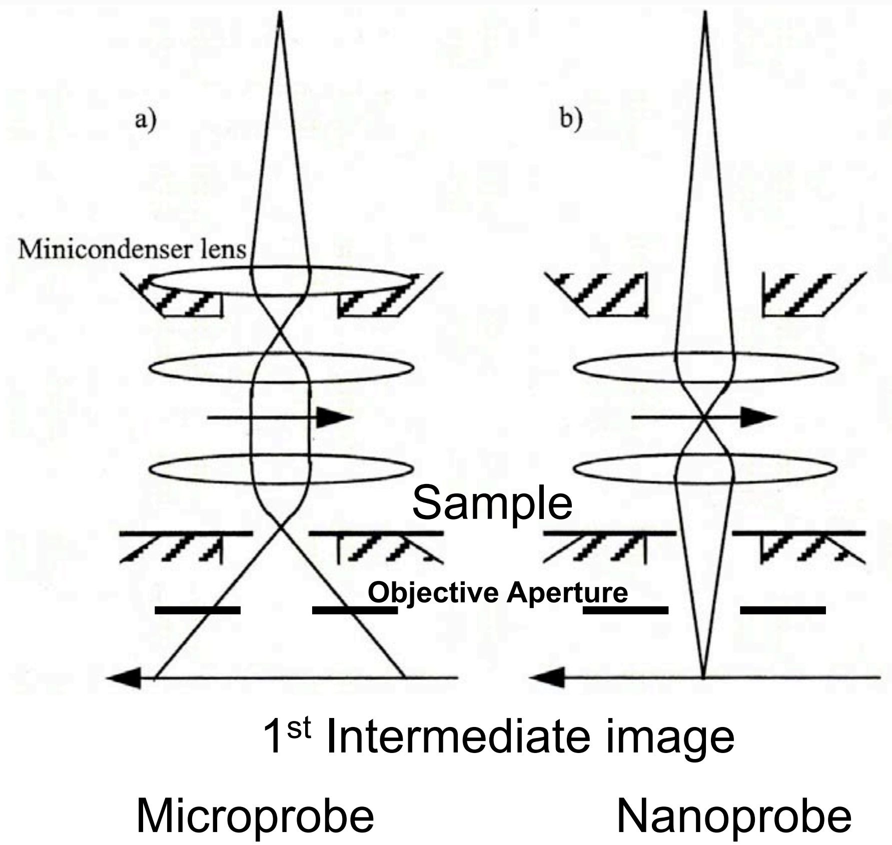
Double Condenser

- With only a single condenser lens, illumination can be varied by changing lens excitation.
- Minimum spot size is determined by magnification of the virtual source.
- A second condenser lens allows changing of the spot size.
- Smaller C2 apertures provide better coherence and correspondingly better resolution, however, intensity is decreased.

In the F-30 the **Spot Size** selector controls C1 lens excitation (changing the size of the gun cross-over) with larger numbers corresponding to smaller spot size, while the **Intensity** controls excitation (focusing and defocusing) of the C2 lens. In combination they allow formation of electron probes of varying size and intensity.

Objective Lens

- 1st Intermediate image of object will be magnified by down-line lenses.
- Smaller the focal length, the better the resolution (smaller pole piece gap).
- However, small gap limits tilt, sample holder options, and ability to position detectors near sample.
- F-30 S-Twin has intermediate pole piece gap which balances resolution, tilt and analytical capabilities.



Twin Lens has minicondenser that is turned on in **Microprobe** mode (wide field of view and coherent illumination), while it is turned off (field reversed) in **Nanoprobe** mode for fine probe illumination.

Imaging Lens System

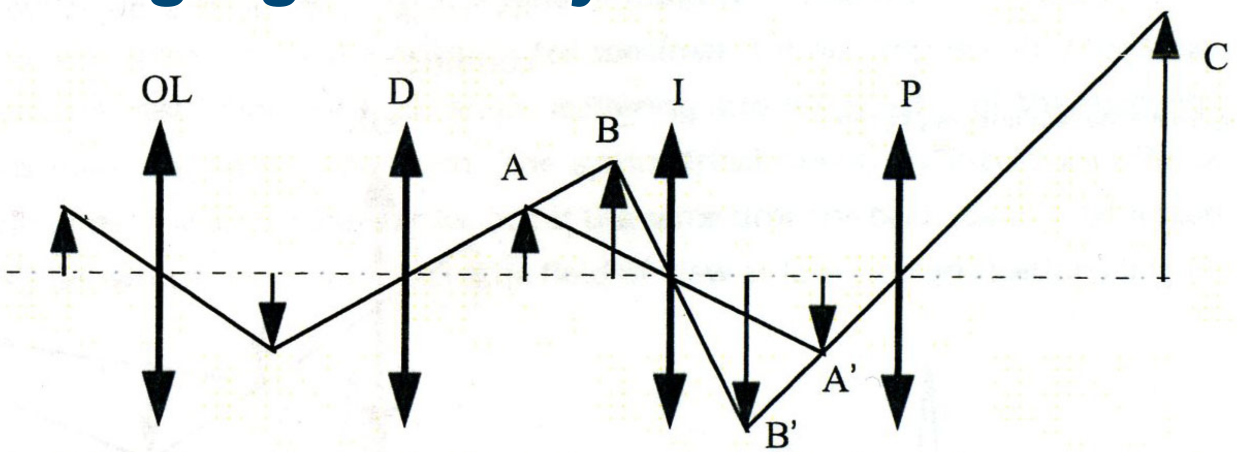
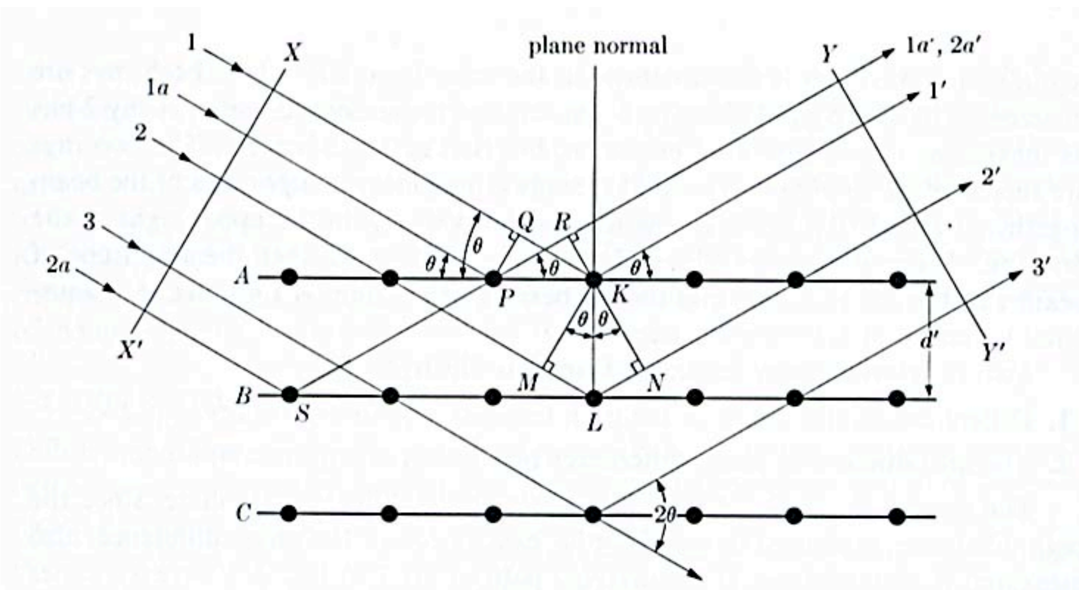


Image plane becomes object plane of next lens. By changing lens excitation, focal length and thus mag. are changed (further from the lens, the greater the magnification).

- Objective lens only magnifies image on the order of 30 x (mag. proportional to focal length).
- Want to be able to have a range of magnifications.
- Also, ideally the image plane needs to remain stationary (no change in Obj. lens current) as magnification is changed
- Requires the use of multiple lenses in imaging system.
- A total of four lenses are used to allow focusing the back focal plan of the Obj. lens (DP) and the image plane over a range of magnifications.
- The image is projected on the photo luminescent (phosphor) screen.
- Due to the nature of the lenses, the depth of focus is very large (on the order of meters) and so multiple imaging detectors can be placed along the optical axis and still have an in focus image (magnification, however, will be different).

Determination of Crystal Structure



Elements of X-Ray Diffraction, B. D. Cullity

Electromagnetic radiation and energetic particles whose wave lengths are on the order of the atomic spacing will scatter when passing through the sample.

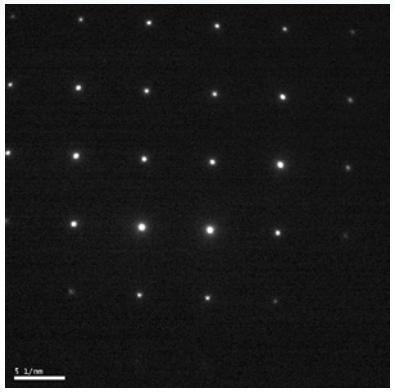
In a crystalline materials, the wave nature of the electron requires that scattering behavior of an electron with wavelength λ is governed by Bragg's law:

$$n\lambda = 2d \sin\theta$$

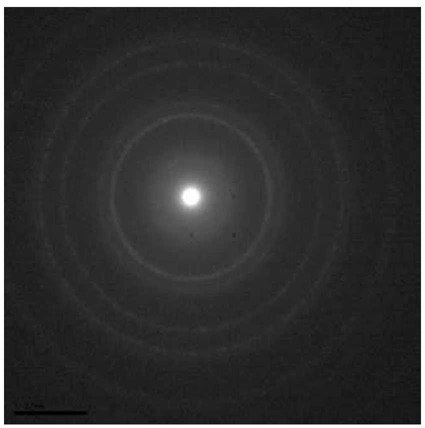
where θ is the scattering angle, n is an integer multiple and d is the planar spacing. Only electrons satisfying Bragg's law will constructively interfere, all others will destructively interfere.

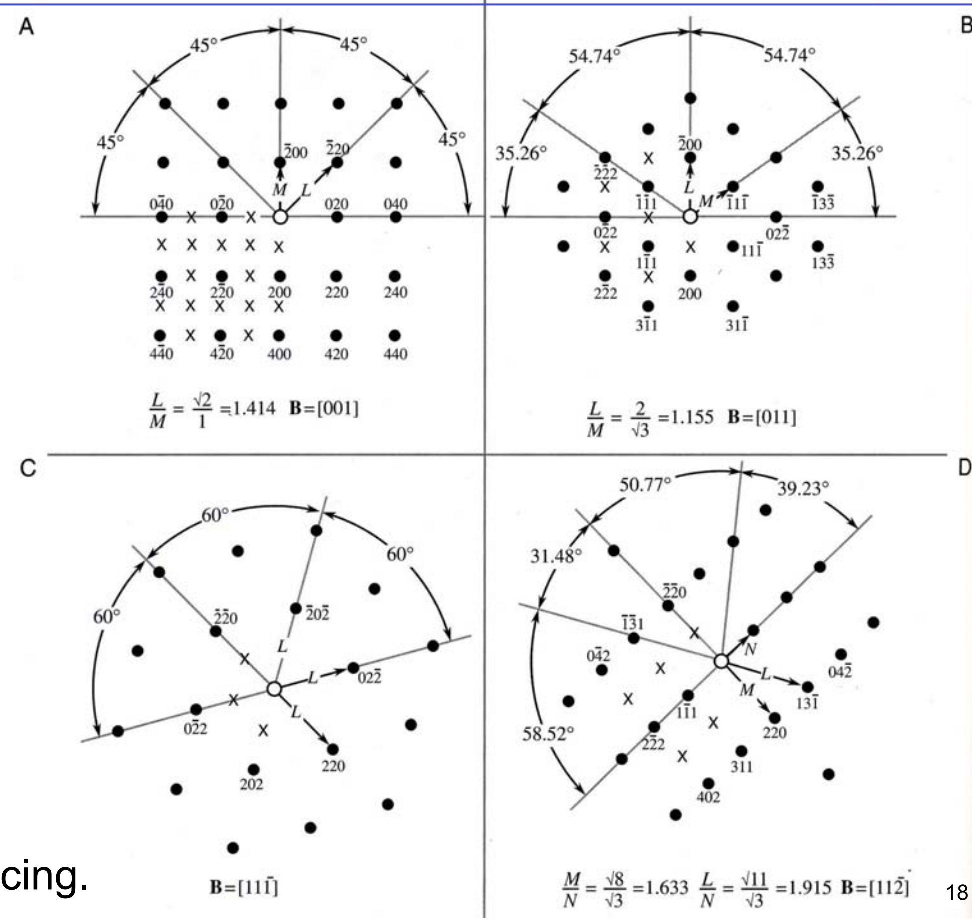
The consequence of Bragg's law in the TEM is that when the electron beam passes through the sample, it will be scattered into multiple beams, and a diffraction pattern will be formed in the back focal plane of the objective lens.

Selected Area Diffraction (SAD)



Beam is spread out (<u>Intensity</u> dial) forming a parallel probe (TEM mode). Results in diffraction spots.

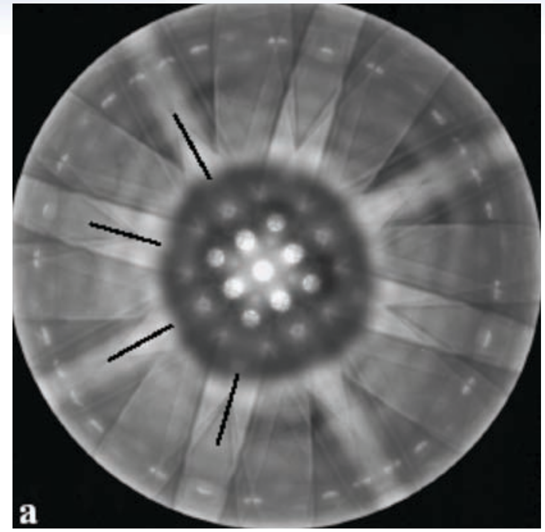


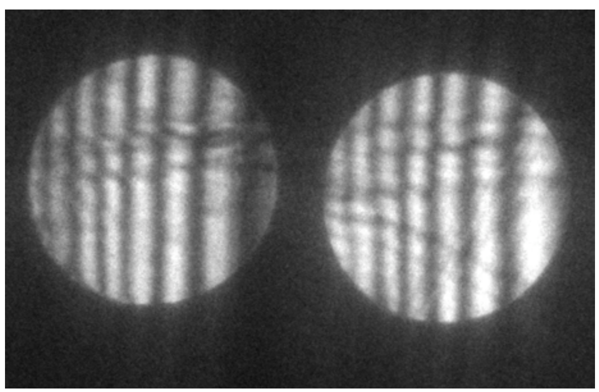


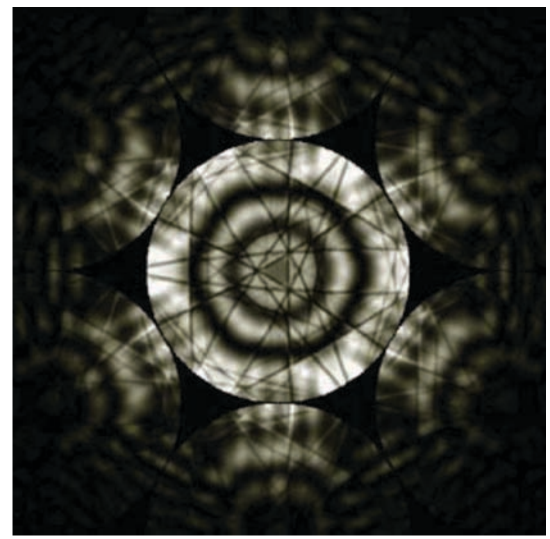
- SAD aperture placed over area interest.
- Single crystal gives spot pattern.
- Polycrystal gives rings.
- Spot spacing inversely related to planar spacing.
- High symmetry patterns indicate low index zone axis.

Convergent Beam Diffraction (CBED)

- Beam is converged to a small probe.
- Diffraction Discs are formed.
- CBED provides additional crystallographic information over SAD including crystal groups.
- Fine structure in central beam can be used to more accurately determine lattice parameter changes.
- Sample thickness can be determined using two beam CBED fringe patterns.

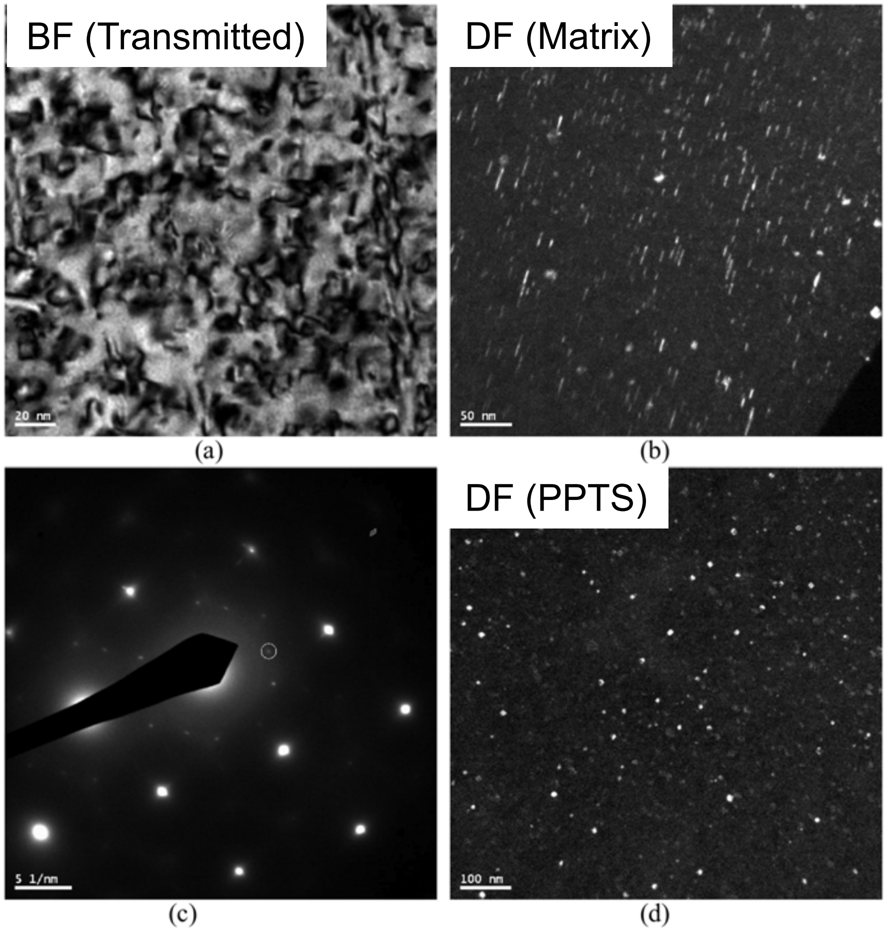






Diffraction Contrast Imaging

- Contrast is provided by forming the image with either the transmitted or one of the scattered electron beams and excluding the others using the objective aperture.
- By imaging with different reflections (spots) present in the diffraction patterns, various features of the microstructure can be revealed in isolation.
- There are two ways these images can be formed.
  - Moving the objective aperture off the optical axis to surround the reflection of interest (poor resolution).
  - Use the beam tilts to bring the diffraction spot on axis.



Defect Analysis in the TEM

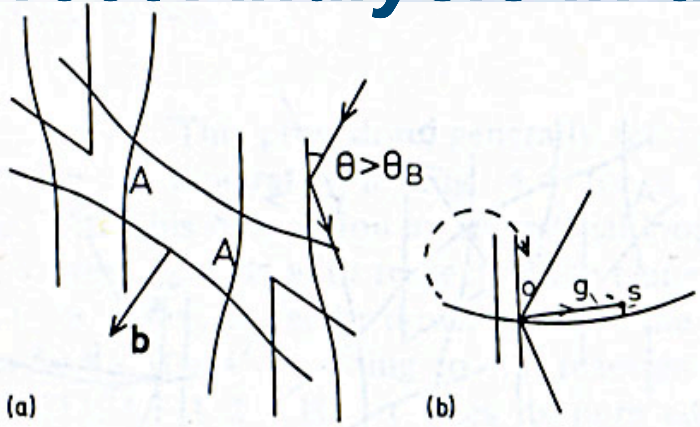
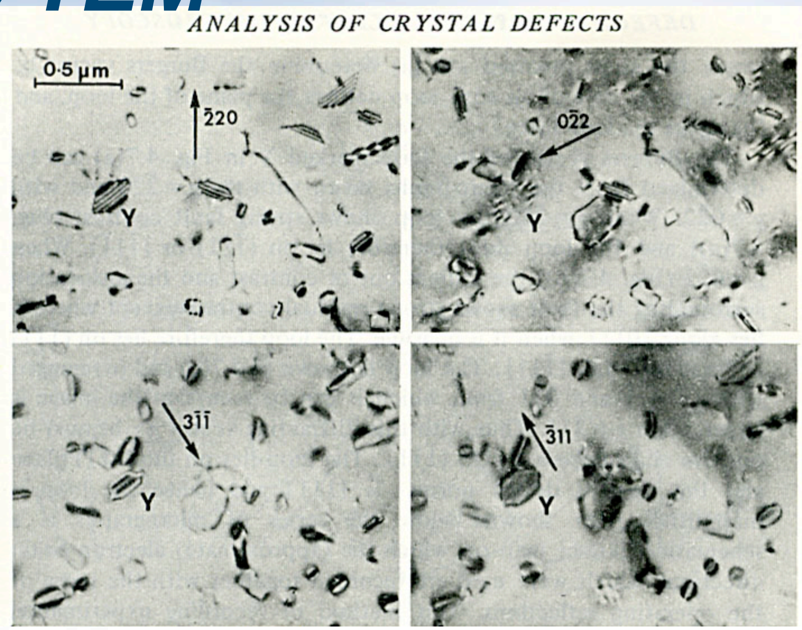
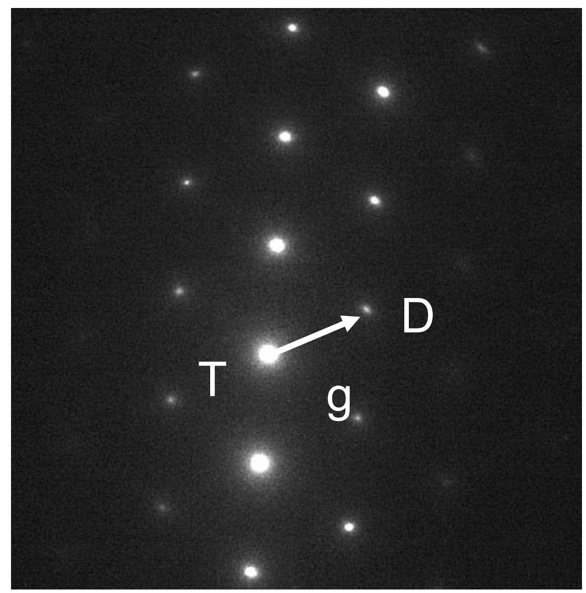


FIG. 4.6

(a) Schematic picture of the sense of the curvature of diffracting planes around a vacancy loop. The diffracting planes are at an angle $\theta > \theta_B$ to the incident electron beam except at A where $\theta \simeq \theta_B$ so that the image of the loop is formed on that side of the core of the dislocation corresponding to A. (b) The sense of g is defined for (a) using the Ewald sphere construction, and the dashed arrow indicates the sense of rotation of the reflecting planes which would lead to strong contrast.

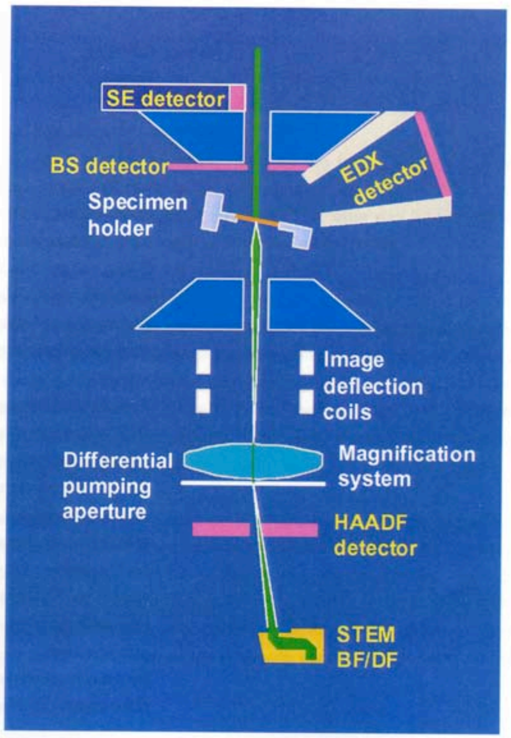




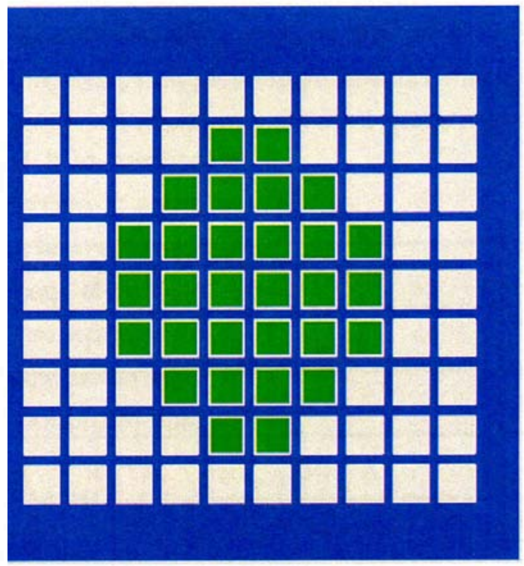
In the FCC crystal, edge dislocations will be invisible when g·b is equal to zero.

Defect analysis in electron microscopy
by M. H Loretto

STEM Imaging



Possible detectors used in STEM



Schematic of point by point signal generation

- Fine probe is scanned over sample in a rectangular pattern using the image deflection coils.
- The detector reading the signal converts the intensity into a brightness value.
- A synchronized pixel by pixel image is created on the viewing monitor that corresponds to the scanned intensity.
- Both images and compositional maps can be produced based on the particular detector acquiring the signal (BF/DF, HAADF, EDX, GIF)
- BF/DF detectors display mainly diffraction contrast (elastic scattering).
- HAADF detector displays elemental or z-contrast (inelastic scattering).

High Resolution Imaging

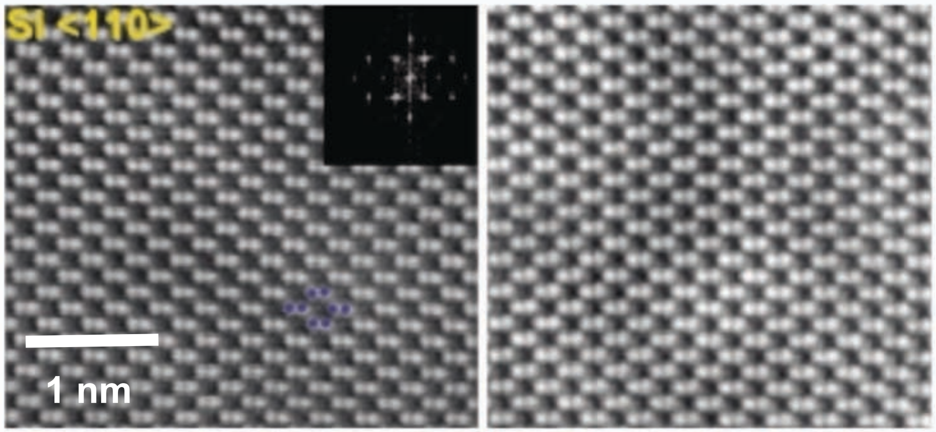


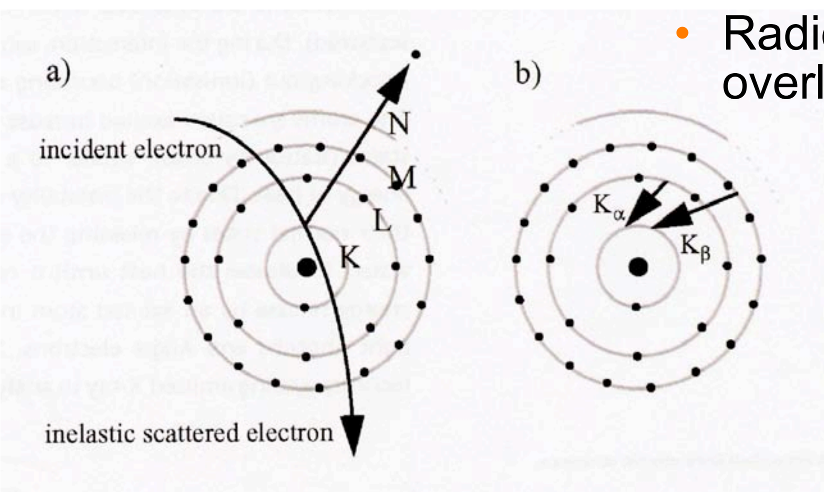
Fig. 1. HR-STEM and HR-TEM image of silicon <110>. In both images the dumbbell structure can clearly be resolved. The atomic positions are marked with the blue dots in the STEM image.

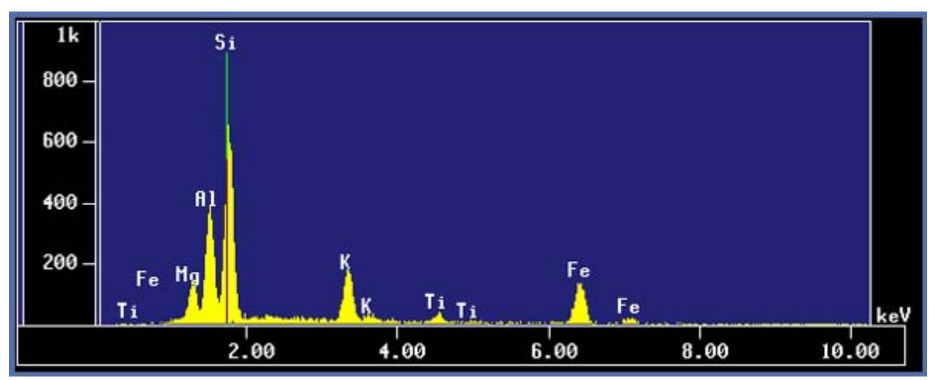
Sample is tilted to zone axis. Microscope is aligned and objective lens stigmation is corrected (no objective aperture used). Focus adjusted until lattice fringes are observed.

- High resolution imaging can be performed both in TEM mode and STEM mode and permits observation of atomic columns.
- Uses phase contrast instead of diffraction contrast (multiple diffracted beams interact to form the image).
- Very thin sample is needed to correctly interpret the image generated.
- Microscope alignment is critical for obtaining good HR images.
- Useful for examining crystal defects such as dislocations and stacking faults as well as interfaces such as grain boundaries.

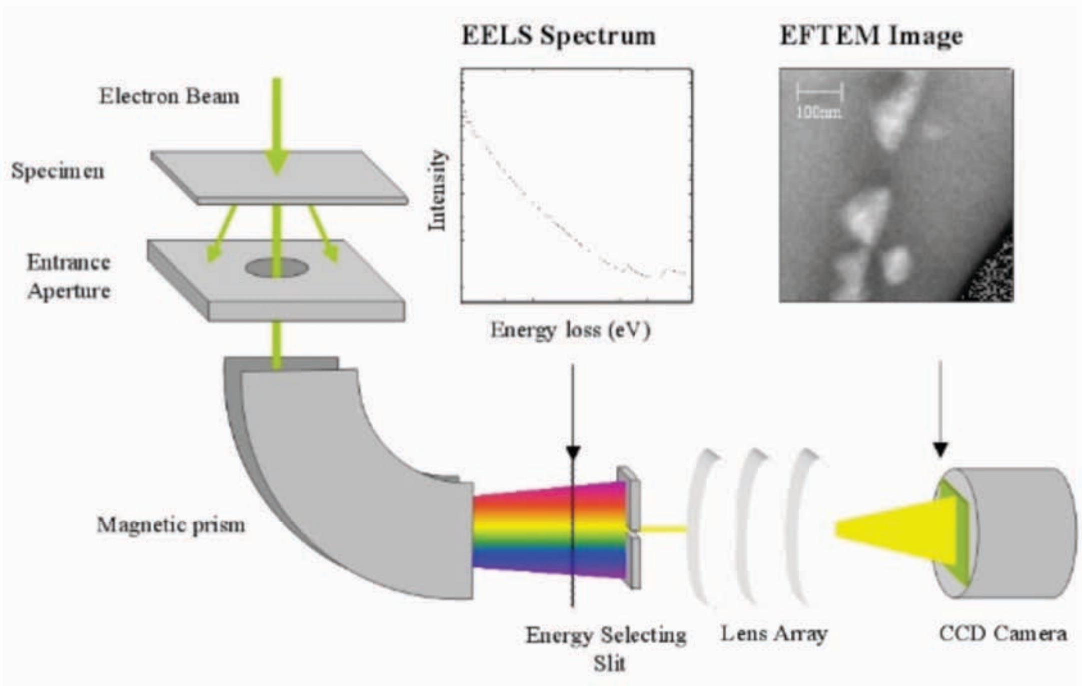
Energy Dispersive Spectroscopy (EDS)

- Incident electron transfers enough energy to inner shell electrons in sample atoms to cause them to be ejected from the atom creating an ionized atom and leaving a hole in the inner shell.
- Outer shell electron drops into hole and emits an x-ray.
- The x-ray energy is characteristic to the emitting atom.
- By measuring emitted x-ray counts as a function of energy, a spectra is generated.
- Semi-quantitative elemental concentrations can be derived from such a spectrum.
- Radioactive materials can also generate x-rays and overload the detector.





EELS and Energy Filtered TEM (EFTEM)

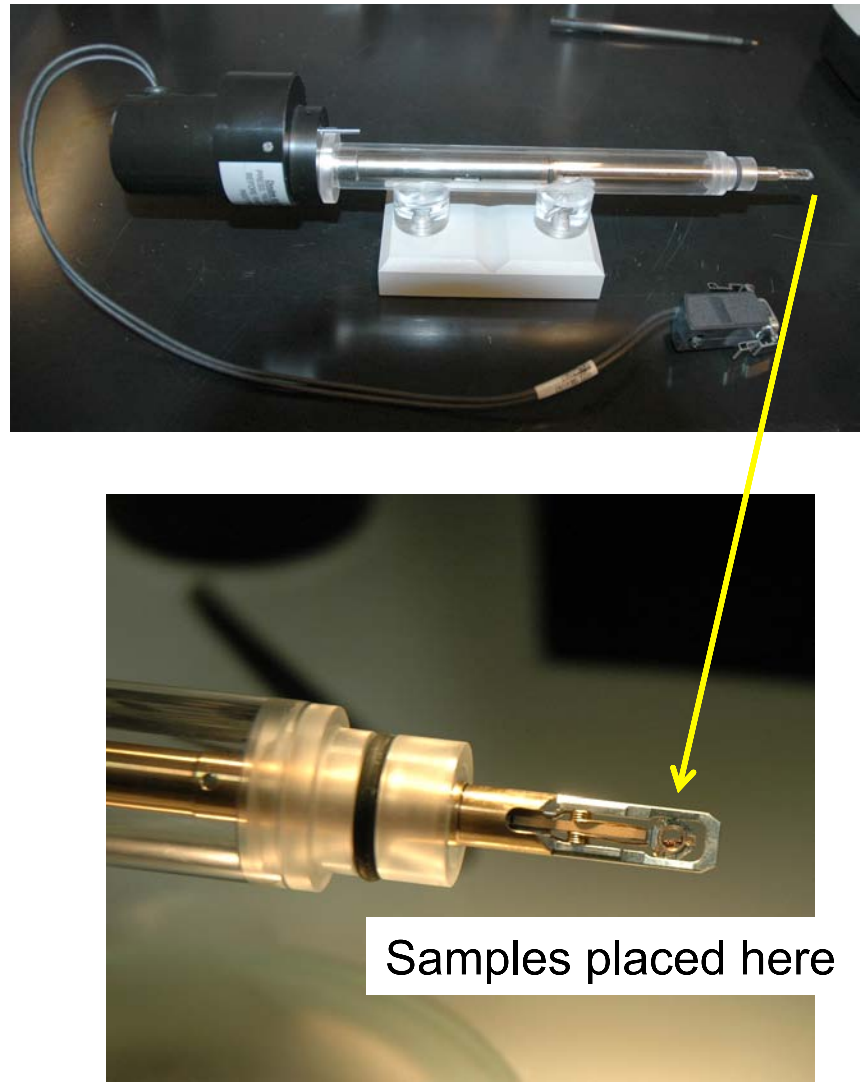


A sector magnet acts like a prism and separates electrons spatially according to their energy. A slit filters the electrons and subsequent lenses restore the original image.

- Electron energy loss spectroscopy (EELS) analyzes the energy distribution of electrons that have undergone inelastic scattering.
- Inelastic scattering is highly dependent on electronic structure (bonding, nearest neighbors etc.)
- In EFTEM, electrons of a particular energy loss are allowed to pass through a slit while others are filtered out.
- EFTEM enables high spatial resolution chemical information to be obtained.

Sample Preparation

- One of the most significant challenges of TEM analysis is preparation of artifact-free, electron transparent samples for analysis.
- TEM samples are typically prepared through a series of sectioning, grinding and polishing steps.
- Metallic samples can be prepared by electrochemical polishing.
- Ceramic samples must be mechanically ground and thinned to electron transparency using ion milling.
- The FIB is can be used to prepare site specific TEM samples.



Sample Preparation Instruments and Technique

Dimple Grinder

Wedge Polisher

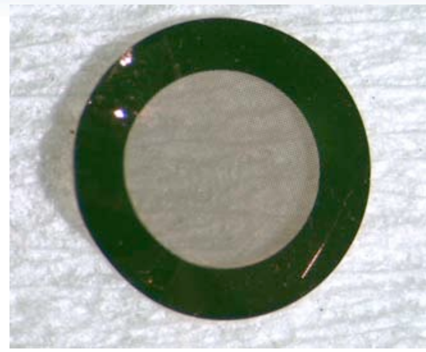
Typical Sample
Profile

Ion Mill

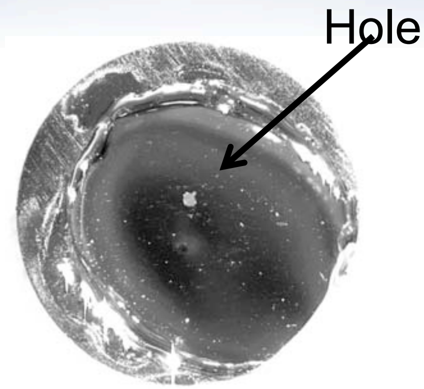
Focused Ion Beam

Electropolisher

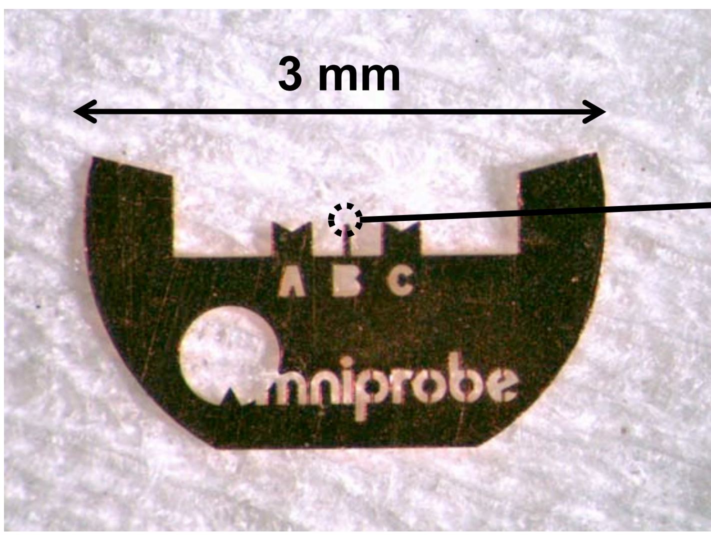
Some Sample Types



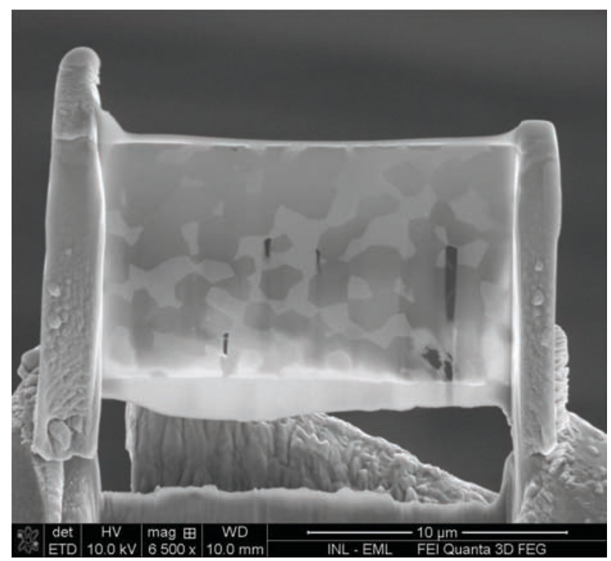
Mesh Grid



Electropolished
Metal Foil



FIB Lift-out Grid

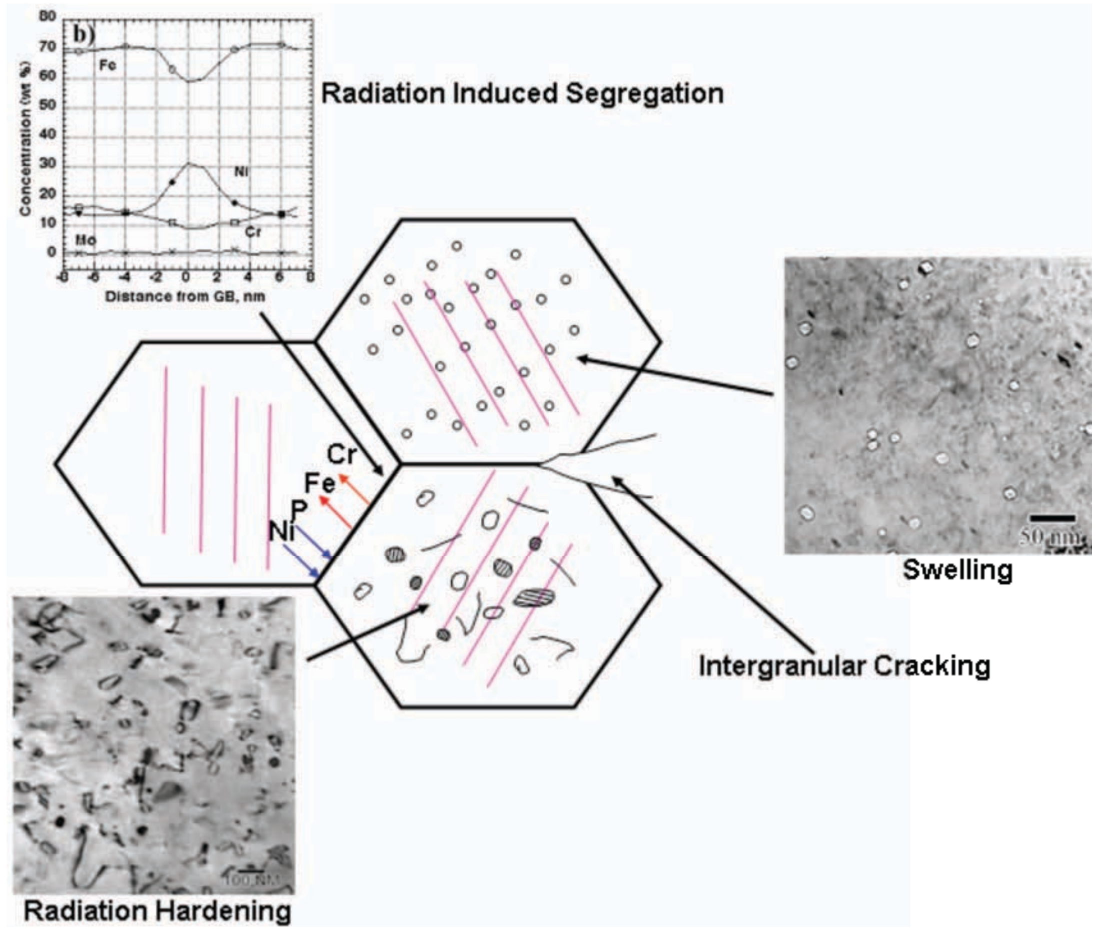


Idaho National Laboratory

Nuclear Fuels and Materials Examples

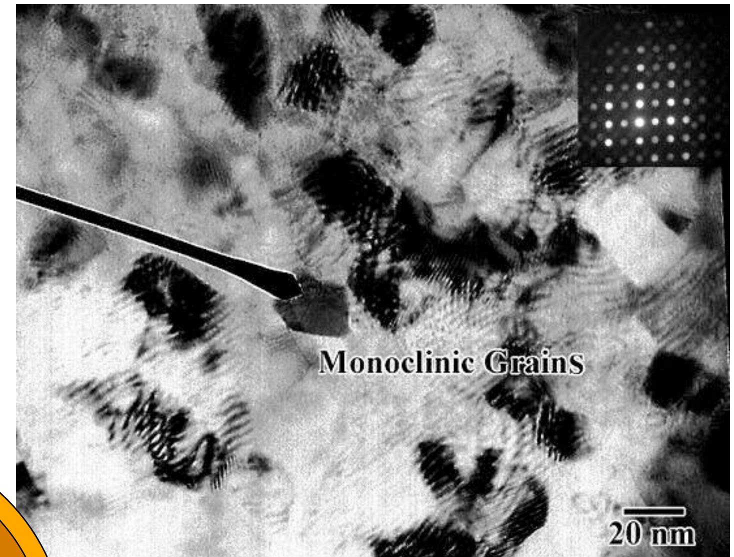
Materials Degradation Phenomena in Austenitic Stainless Steel

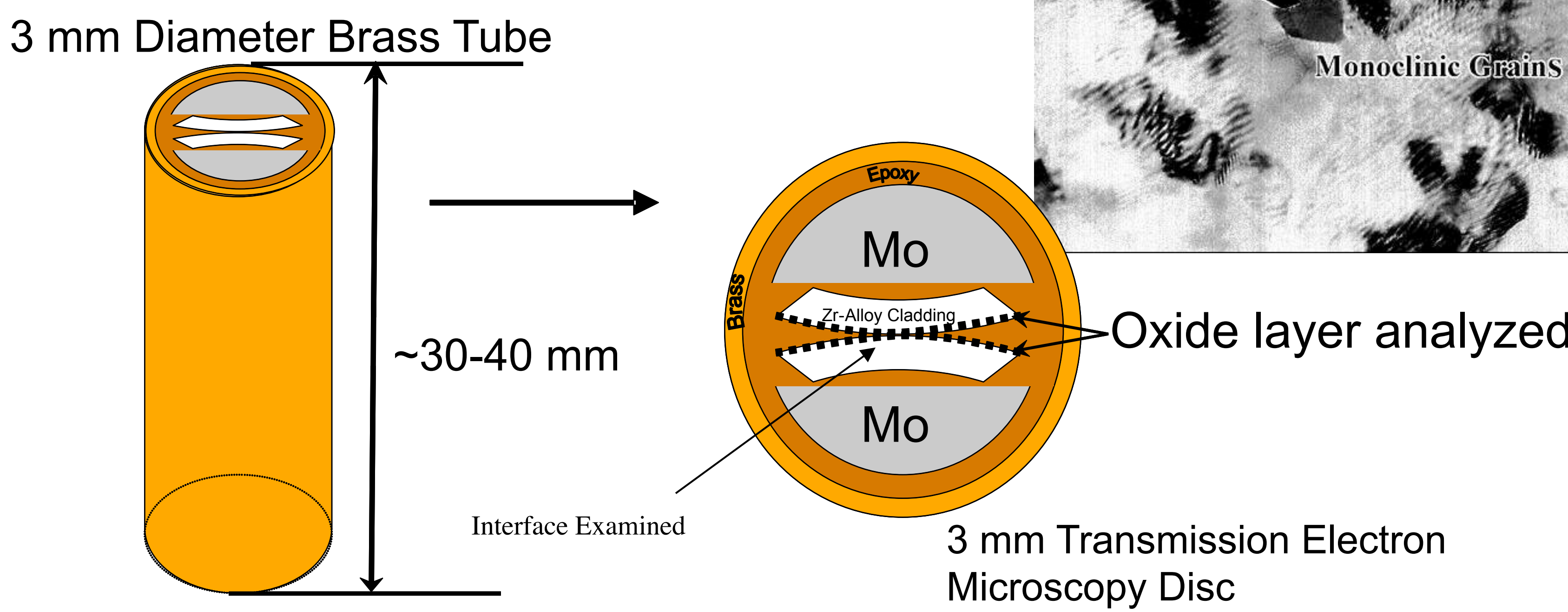
- Development of dislocation and void structures
- Development of radiation induced segregation
- Radiation-induced phase stability
- Radiation-induced dimensional change



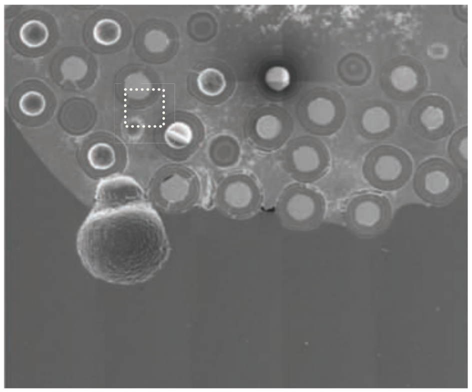
Corrosion of Advanced Zirconium Alloys

- Transmission electron microscopy of irradiated Zr alloys
  - Zry-4, Imp Zry-4, and ZIRLO
- Oxide layer characterization
  - Grain size, morphology, Second Phases

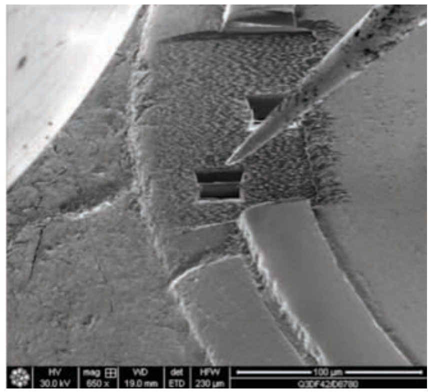




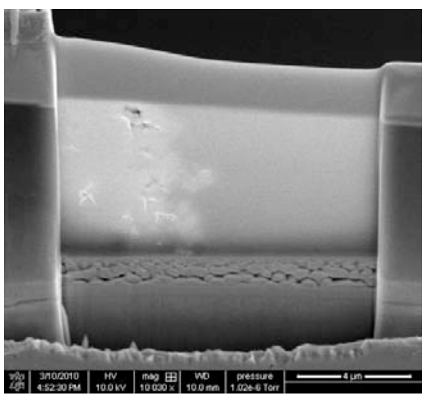
FIB Lift-Out of Surrogate Triso Fuel Particle Interface Sample



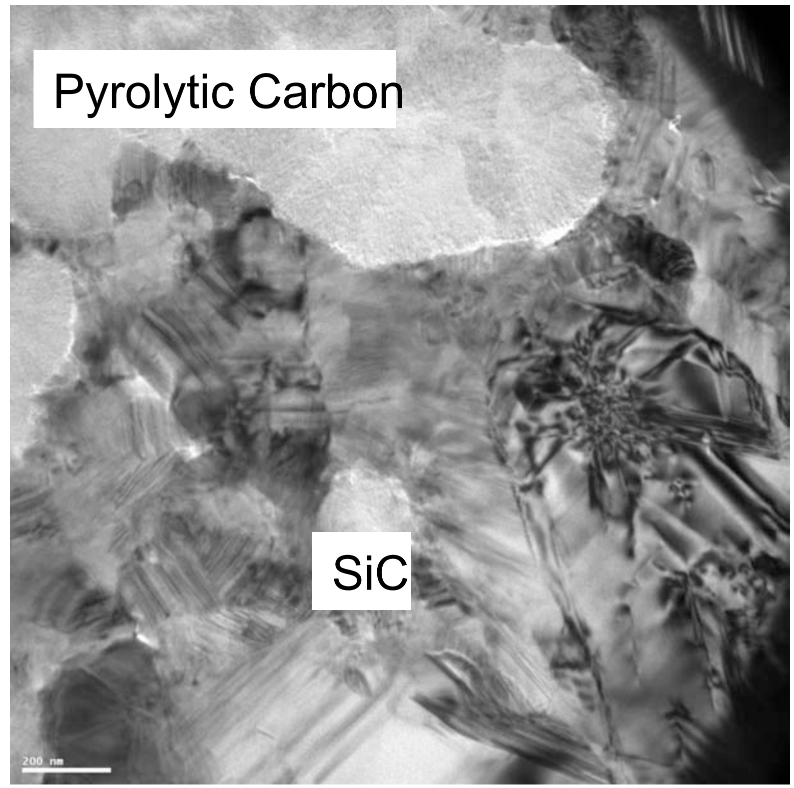
Triso particles with surrogate ZrC fuel kernel



Liftout of interface sample



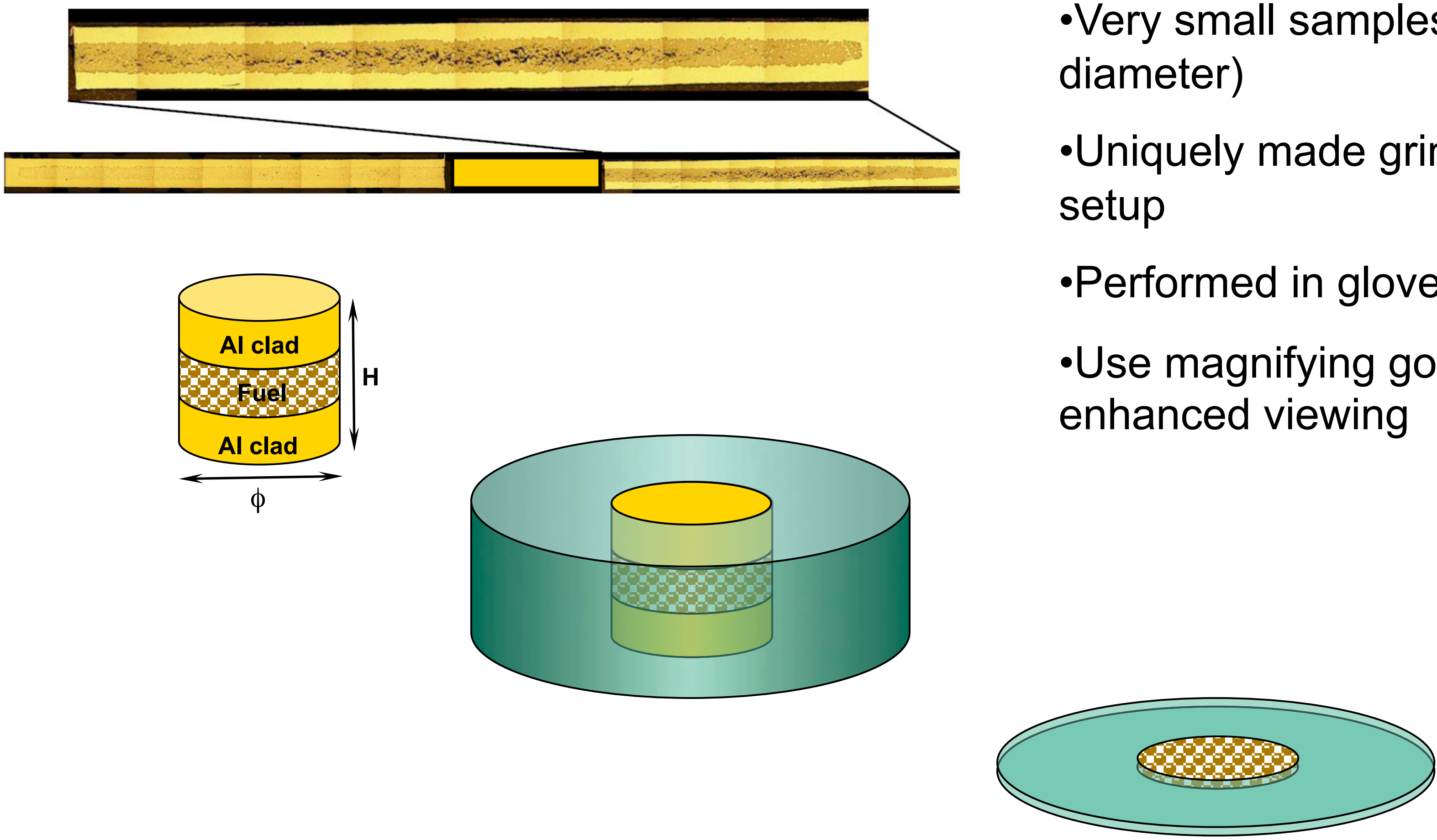
Thinned TEM lamellae



TEM image of interface

Courtesy of R. Kirchhofer

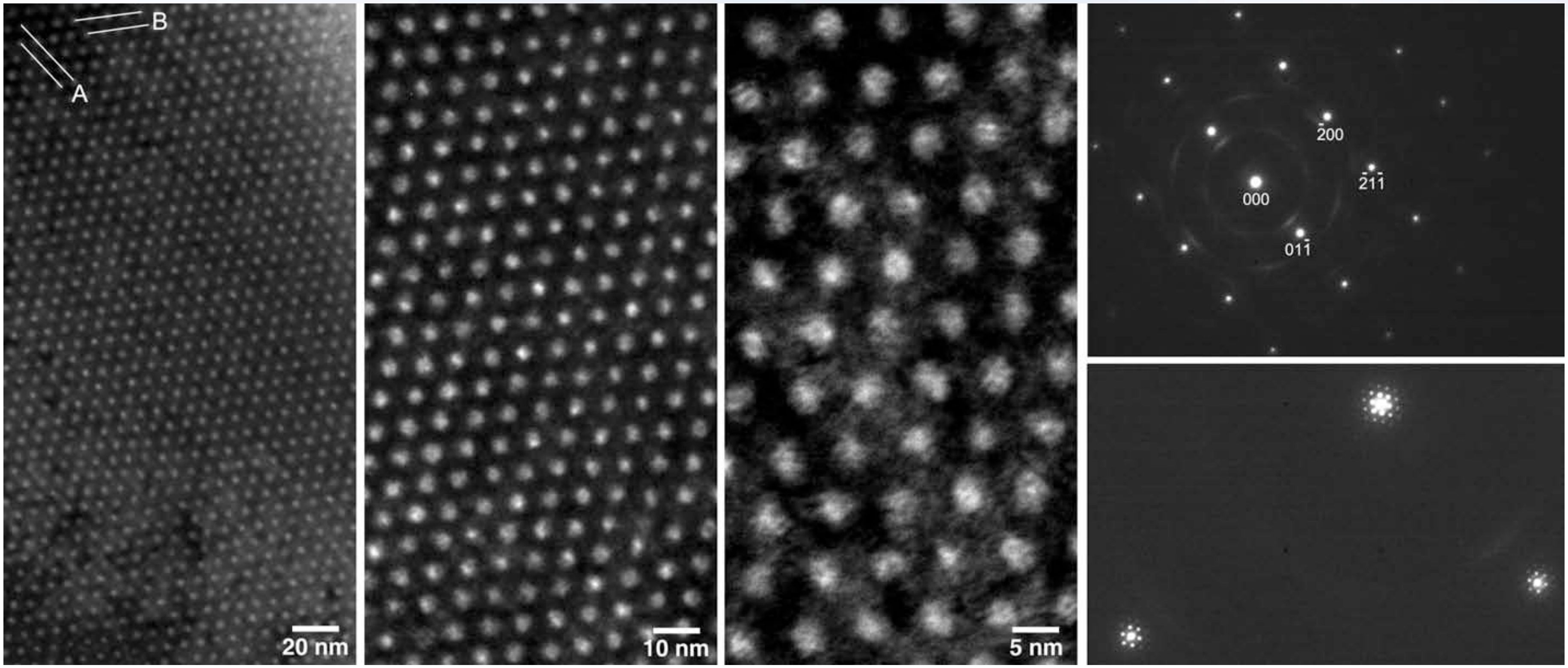
TEM Sample Prep of Irradiated Plate Fuel



- Very small samples (1mm diameter)
- Uniquely made grinding setup
- Performed in glove box
- Use magnifying goggles for enhanced viewing

Courtesy of Jian Gan

Superlattice Bubbles in Irradiated U-7Mo Particle



- TEM images showing the superlattice of fission gas bubbles observed in a U-7Mo particle of bcc structure orientated at zone [011].
- The selected area diffraction (SAD) patterns showing rings due to oxides formed at the U-7Mo fuel particle from sample preparation.
- An enlarged view of the SAD pattern showing the satellite spots due to the bubble superlattice.

Courtesy of Jian Gan

Selected References

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- M. H. Loretto, Electron Beam Analysis of Materials, 1984, Chapman and Hall, New York.
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